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Original Article

Fabrication and properties of dense α-cordierite glass-ceramic coating on porous BN/Si_2N_2O ceramic

Yangshan Sun[a,b], Delong Cai[a,b], Zhihua Yang[a,b,c,*], Hailiang Li[a,b], Bo Niu[a,b], Wenjiu Duan[a,b], Dechang Jia[a,b,c,*], Yu Zhou[a,b]

[a] Institute for Advanced Ceramics, School of Materials Science and Engineering, Harbin Institute of Technology, Harbin, Heilongjiang, 150080, China
[b] Key Laboratory of Advanced Structrual-Functional Integration Materials & Green Manufacturing Technology, Harbin Institute of Technology, Harbin, Heilongjiang, 150001, China
[c] State Key Laboratory of Advanced Welding and Joining, Harbin Institute of Technology, Harbin, Heilongjiang, 150001, China

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ABSTRACT

α-Cordierite glass-ceramic coating was fabricated on the porous BN/Si_2N_2O ceramic by glass-ceramic method. The effect of the heating temperature on the phase composition, microstructure, mechanical properties, water resistance and dielectric properties of the coatings was investigated. A large amount of α-cordierite precipitated from the glass phase when the heating temperature was 1050 °C and the content of α-cordierite in the coating increased with increasing the heating temperature. The resulting α-cordierite glass-ceramic coatings had a good wettability and adhesion with the porous ceramic substrate. The coating/substrate interface was continuous without defects. When the heating temperature was 1050–1200 °C, the resulting coatings possessed positive mechanical properties and good water resistance due to the high densification. And the dielectric constant and loss tangent of the coated samples prepared at 1050–1200 °C were 4.1–4.3 and 0.005–0.01 respectively in the frequency of 21–36 GHz.

1. Introduction

The porous boron nitride/silicon oxynitride (BN/Si_2N_2O) ceramic is a promising candidate material for high temperature wave transparent applications owing to its good dielectric properties ($\varepsilon < 4.49$, $\tan\delta < 0.0056$), high temperature stability, positive machinability, low density and great thermal shock resistance [1–4]. The dielectric properties and thermal insulation of the porous BN/Si_2N_2O ceramic can be improved by increasing porosity [5,6]. Unfortunately, the porous BN/Si_2N_2O ceramic with many pores can easily absorb moisture in the air, resulting in a decrease of dielectric properties and thermal insulation [7,8]. Moreover, high porosity of the porous BN/Si_2N_2O ceramic can lead to the degradation of rain and particle erosion resistance and mechanical properties [9]. Consequently, the above problems will limit the application of the porous BN/Si_2N_2O ceramic in radome and antenna window.

Therefore, it is critical to apply a dense wave transparent coating on the surface of the porous BN/Si_2N_2O ceramic in order to solve its problems mentioned above. In recent years, cordierite based glass-ceramics are prime candidates for the protective coating of C/C and SiC_f/SiC composites due to their low thermal expansion coefficient, high thermal and chemical stabilities [10–12]. Moreover, it is well known that α-cordierite glass-ceramic possesses low dielectric constant and dielectric loss [13–16], which is essential for its use as a wave transparent coating material. Therefore, through the above discussion, α-cordierite glass-ceramic is a very competitive candidate material for a dense wave transparent coating on the surface of the porous BN/Si_2N_2O ceramic.

Up to now, there are several feasible methods for fabricating α-cordierite based glass-ceramic coating on the ceramic substrate, such as plasma spraying [17], chemical vapour deposition (CVD) [18] and liquid sintering [19,20]. However, the porous surface of BN/Si_2N_2O ceramic will be damaged by mechanical erosion during plasma spraying process due to the formation of high velocity flight droplets (400 ~ 800 m/s) [21]. And plasma sprayed coatings have intrinsic microcracks and porosity. The pure and crystalline coating can be obtained by CVD method, but the deposition rate is low and some cracks are also present in the CVD coating [18]. Liquid sintering method is very effective to prepare dense coatings, but the shrinkage of the coating is large. As a consequence, stress cracks are generated inevitably in the coating because of the mismatch of shrinkage between the substrate and coating [22,23]. At present, little research has been done about fabrication of α-cordierite glass-ceramic coating on the porous BN/Si_2N_2O ceramic.

* Corresponding author at: Building C3, No. 2 Yikuang Street, Nangang District, Harbin, 150001, China.
E-mail addresses: zhyang@hit.edu.cn (Z. Yang), dcjia@hit.edu.cn (D. Jia).

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In this work, a MAS (MgO-Al_2O_3-SiO_2-B_2O_3) based glass is proposed to fabricate a dense α-cordierite glass-ceramic coating on the porous BN/Si_2N_2O ceramic using glass-ceramic method [10,24]. In this process, heating temperature is a crucial factor for microstructure evolution, crystallization and densification of the coating. Thus, the effect of the heating temperature on the phase composition, microstructure, mechanical properties, water resistance and dielectric properties of the α-cordierite glass-ceramic coating was investigated in detail.

2. Experimental

2.1. Preparation of the coating

The porous BN/Si_2N_2O ceramic composite used as substrate was prepared by pressureless sintering in our previous work [4]. The median pore diameter and open porosity of the porous BN/Si_2N_2O ceramic are about 0.7 μm and 30.4%, respectively. And its phase compositions are Si_2N_2O and h-BN together with a small amount of β-Si_3N_4. The porous BN/Si_2N_2O ceramic was machined into disk samples with dimensions Φ50 mm × 3.5 mm, then ground and polished with 0.4 μm SiC abrasive paper, ultrasonic cleaned with ethanol and dried in drying oven.

MAS based glass was used as coating material, which is based on 20 g MgO powder (purity > 98%, d = 5.83 μm), 24 g Al_2O_3 powder (purity > 98%, d = 0.73 μm), 56 g fused quartz powder (purity > 98%, d = 3.39 μm), and 15 g B_2O_3 powder (purity > 98%, d = 74 μm). Powders of oxides were mixed together in a corundum crucible and melted at 1500 °C for 2 h in a muffle stove, then quenched in distilled water to form glass frits. After that, the glass frits were milled in alcohol at 500 rpm for 2 h, then dried and screened through a 200 mesh sieve. The compositions of the glass powders were analyzed using inductively coupled plasma optical emission spectrometer (ICP-OES, Optima 5300DV, PerkinElmer), as listed in Table 1. The slurry compositions of coating were 70 wt% glass powder and 30 wt% organic solvents, and mixed evenly by ball-milling for 30 min. The organic solvent was composed of 81 wt% solvents (butyl carbitol), 10 wt% plasticizers (tributyl citrate), 5 wt% surfactants (sorbitan trioleate) and 4 wt% adhesives (ethyl cellulose). Then the slurry was spread on the porous BN/Si_2N_2O ceramics surface by a scraping method and decomposition of organic solvents was conducted in a muffle oven at 500 °C for 2 h in air with a heating rate of 0.5 °C/min. The heating temperature and holding time were selected in range of 950–1300 °C and 3 h, respectively.

2.2. Characterization and tests

The phase compositions of the coating were analyzed by X-ray diffraction (XRD, D/max-γB, Ricoh). The natural surface and polished cross-section morphologies of the coating were characterized with scanning electron microscopy (SEM, HELIOS NanoLab 600i), equipped with energy dispersive spectroscopy (EDS). The FIB slice was prepared by HELIOS Nanolab600i equipment. The microstructure of the interface between the coating and substrate was observed by transmission electron microscope (TEM, 200 kV, Talos F200x), and energy dispersive spectroscopy (EDS) analysis was conducted by high angle annular dark field (HAADF), high-resolution TEM (HRTEM), and selected-area electron diffraction (SAED) techniques.

The water absorption and apparent porosity of the porous BN/Si_2N_2O ceramic with and without coatings were assessed by the Archimedes method using distilled water as the medium. The hardness and elastic modulus of the coating were measured by Nano-indentation test, which was performed by Nano-Indenter (Nano-Indenter G200) on the polished cross-section of the coatings. All of the measurements were made under a constant load value of 98 mN with a dwell time of 10 s. The adhesive strength of the coatings was measured using a universal testing machine (Instron-5569, Instron Group) applying a crosshead speed of 0.5 mm/min according to the GB8642-88 method. Dielectric constants and loss tangents of the porous BN/Si_2N_2O ceramic with and without coatings were carried out using RF impedance/material analyzer (PNA N5230 A, Agilent) over 21 ~ 36 GHz at room temperature (RT), the specimen sizes were Φ18.0 mm × 1.0 mm.

Table 1
The compositions of the glass powders.

Element	Mg k	Al k	Si k	O k	B k
Wt.%	11.66	13.65	32.11	41.62	0.96

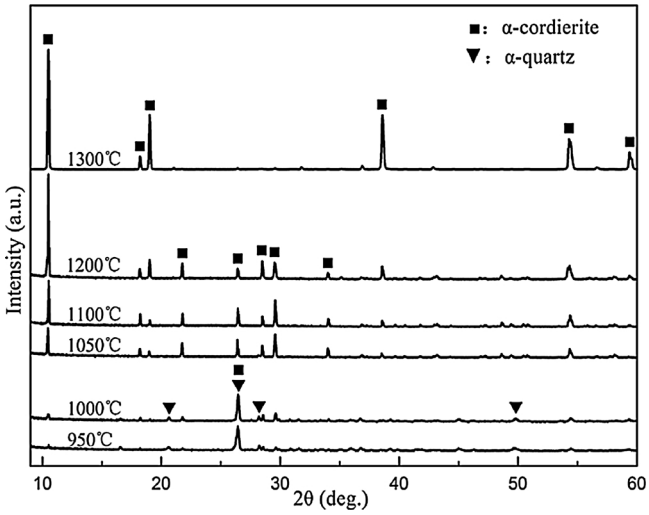

Fig. 1. XRD patterns of the coatings prepared at different heating temperatures.

3. Results and discussion

3.1. Phase assemblages and microstructures

Fig. 1 shows the phase evolution of the coatings fabricated at different heating temperatures. The phase compositions of the coatings consisted of a dominant glass phase with a minor α-quartz and α-cordierite phase when the heating temperature was below 1050 °C. With the increase of the heating temperature, α-cordierite became the dominant phase as a large amount of α-cordierite precipitated from the glass phase. This was because increasing the heating temperature can reduce the viscosity of the glass phase and facilitate substances transfer [25], which can contribute to the crystallization of Mg-Si-Al-O liquid. Meanwhile, the α-quartz phase disappeared and the glass phase decreased gradually with the increase of the heating temperature.

Figs. 2 and 3 show the SEM surface and cross-section images of the coatings prepared at different heating temperatures. It was obvious that large amounts of pores were formed within the coating when the coatings were fabricated at 950 and 1000 °C. The density of the coating increased gradually with increasing the heating temperature, which should be attributed to the reduced viscosity of the glass phase. The glass viscosity-temperature relations can be expressed by the Vogel-Fulcher-Tammann function (1) [26,27],

$$\lg\eta = A + B/(T - T_0) \quad (1)$$

Where η and T are the viscosity and temperature respectively, A, B and T_0 are constants determined by

$$A = 1.455 + \sum a_i[p_i] \quad (2)$$

$$B = 5736.4 + \sum b_i[p_i] \quad (3)$$

$$T_0 = 198.1 + \sum t_i[p_i] \quad (4)$$